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EFFICIENT DETECTION OF LOW LEVELS OF RADIOACTIVITY IN SEAWATER

by

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ADMINISTRATIVE INFORMATION

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ABSTRACT

The problem of assaying radioactivity in the sea has been under study with a view to designing an optimum detector for gamma radiation spectra, with suitable statistical accuracy.

The responses of a series of crystal detector spheres of radii from 0.1 cm to 15 cm, having unit intrinsic efficiency to gamma rays has been calculated for unit gamma emitter per unit volume. The principal result brought out is the large increase in counting rate as the solid angle of acceptance of the spheres for gamma radiation increases. With larger spheres the sample volume increases but gives a negligible increase in counting rate. Graphs are given for the series of spheres for energies between 0.137 Mev and 3.6 Mev. A formula is given for the maximum counting rate for any size sphere under bombardment with any gamma ray energy. It can be used to estimate the size detector needed to count an emitter with required statistical accuracy, making allowance for intrinsic efficiency of the detector to gamma rays of a particular energy, and for any concentration of emitter.

A procedure is outlined for selecting the particular isotope which has been activated by unit neutron fluence to obtain the maximum counting information in the shortest time. Differential counting of two isotopes over two different periods of time should give information on recency of activation.

SUMMARY

The Problem:

To optimize the size of gamma radiation spectral detectors for oceanographic measurements.

The Findings:

In a distributed gamma radiation source of infinite extent, the detection rate of a crystal is proportional to the square of the detector radius, and increases with the energy of the primary gamma ray.

For a given detector size, the response approaches a limit with increasing source volume.

With lower energy gamma rays, the efficiency of a detector per unit volume of crystal may be low, because of attenuation of the rays in the crystal surface-layers. A group of smaller detectors, set at about two absorption lengths of the absorbing medium from each other, may therefore give a considerably higher count rate than the same volume of crystal in a single piece.

A procedure is suggested for selecting the time of assay for a particular isotope in a mixture of activities (such as fission products in water) having a continuous distribution of mean lifetimes. At this optimum time (which is the mean lifetime), interference from shorter-lived activities is least. Other procedures are discussed for selecting a particular isotope to be assayed in a discrete distribution of activities of various mean lifetimes.

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INTRODUCTION

The problem of assaying radioactivity in the sea has been under study with a view to designing a practical detector capable of giving information on gamma radiation spectra. At very low concentrations of a particular active isotope, when appreciable background from other isotopes exists, the number of counts necessary for a statistically reliable estimate of this particular activity requires considerable time.

Both more sensitive and more efficient detection instruments are needed than those presently available. The problem of increasing the luminescence efficiency of the crystal will not be considered. Its importance, however, is pointed out in directing the assay toward a particular isotope which can be counted efficiently. The principal problem attacked is that of estimating the probability of detecting activity distributed in surrounding seawater and of showing quantitatively the increasing counting rate with increasing solid angle of acceptance of the detector for surrounding activity. A procedure for using these computations for determining which particular radioisotope should be counted is outlined.

In carrying out the analysis, the responses are computed of a series of detecting spheres (thought of as crystals) of increasing radii placed in homogeneous absorbing seawater with unit activity of particular gamma emitter per unit volume. The detector is assumed ideal, of unit intrinsic efficiency, with every gamma ray of a particular isotope reaching the surface from the surrounding seawater being counted and giving an equal signal above instrument threshold. Only primary photons are considered, the response to scattered photons being neglected for present purposes.

An integral equation is developed for the probability of detection of activity in an element of the medium surrounding the detector, in terms of the radius of the detecting sphere, and the transmission of the gamma ray to the sphere through the surrounding medium. Summing the products of these two probabilities throughout the active medium gives the counting rate for unit activity density in the medium and unit efficiency of the detector.

A graph gives the shape of the response curve for any radius of the active medium surrounding the sensitive sphere. The formulas developed have been used to compute responses to activity from close in to several mean free paths for transmission through the absorbing medium. For activity distributions to some distance from the detector, simple asymptotic relations give a good approximation to the maximum counting rate from the distributed source. The results show the very great increase in detection rate with increasing radius of the spherical detector and give a simple relation from which the maximum counting rates for detectors of any radii can be computed.

CALCULATION OF DETECTION RATE

Consider a homogeneous detecting sphere, Fig. 1, of radius a, sensitive to the traverse of gamma radiation and with the center at distance D from an element of radioactivity dV in the surrounding medium. The counting rate N arises from the interception by the sphere of the primary photons which are transmitted through the medium from the volume element.

$$N = \epsilon\rho \int_0^\infty \int_{Sp} \frac{d\Omega}{4\pi} e^{-\mu r} dV \qquad (1)$$

where dV refers to the volume containing radioactivity, $d\Omega/4\pi$ to the elementary fractional solid angle of acceptance on the spherical surface from the element of active volume.

The distance r is that from the volume element of activity to the front face of the detecting sphere, that is, the distance travelled by the ray from element dV to the sphere, and μ is the attenuation coefficient for primary rays of a particular energy. The quantity ϵ is the intrinsic efficiency of the detector material, or counts per photon on the sphere, and ρ the gamma ray source strength per unit volume.

This fundamental form can be looked at as the product of two probabilities summed over the activity distribution: $d\Omega/4\pi$ is the probability that a ray from the volume element will start toward an element of solid angle on the sphere and $e^{-\mu r}$ as the probability that the ray will traverse the medium until it is intercepted by the sphere. A problem remaining is to put these quantities into integrable terms. It is physically meaningful to choose α and D as the independent

variables in which to evaluate the double integral, as in the following.

Probability of Direction.

An element of total solid angle (Fig. 1) subtended on the sphere from the source point P is: $2\pi \sin\alpha \, d\alpha$ and the probability that a ray will start toward the element is

$$\frac{d\Omega}{4\pi} = \frac{2\pi \sin\alpha \, d\alpha}{4\pi}$$

Probability of Transmission.

To evaluate r in terms of D and angle alpha the cosine law is used (Fig. 1):

From this, $a^2 = D^2 + r^2 - 2r\,D\cos\alpha$

Solving for r: $r = D\cos\alpha - \sqrt{a^2 - D^2 \sin^2\alpha}$

So the factor $e^{-\mu r}$ becomes $e^{-\mu(D\cos\alpha - \sqrt{a^2 - D^2\sin^2\alpha})}$ and can be inserted in formula (1) for evaluation.

Volume of Activity.

The remaining quantity to be expressed in terms of D is the volume element dV. Considering a shell of seawater at distance D the volume element is evidently

$$dV = 4\pi D^2 dD.$$

With these several quantities evaluated in terms of D and the angle α the double integration can be performed on a machine between the limits of distance from which appreciable activity can be counted.

The final expression becomes:

$$N = \frac{\varepsilon\rho}{4\pi} \int_a^D dD \int_0^{\sin^{-1}\frac{a}{D}} d\alpha \, 4\pi D^2 \cdot 2\pi \sin\alpha \cdot e^{-\mu(D\cos\alpha - \sqrt{a^2 - D^2 \sin^2\alpha})} \qquad \text{(II)}$$

Before discussing the results for different photon energies and different size spheres a general picture of the results can be seen as follows: Because of the nature of the distributed source around the detector, it is evident that beyond a certain number of mean free paths for transmission for a given energy the remaining source contributes nothing to the counting rate. At the other extreme, with no distributed source beyond the detector, no counts will occur. The vertical asymptote for the lower limit is seen to be D=a, since close in the maximum value of the angle α is $\pi/2$ at which time the exponential factor becomes unity, leaving only the factor the integral of $2\pi D^2 dD$ between a and a. Hence the expression amounts to zero close to the asymptote.

Next, the horizontal asymptote becomes $\frac{\pi a^2 \varepsilon\rho}{\mu}$ when angle α becomes very small and the solid angle of the detector is given by the area of a diametral plane of the detector divided by D^2 where D is the distance from the source point. The expression becomes

$$N = \varepsilon\rho \int_a^D 4\pi D^2 \cdot \frac{\pi a^2}{4\pi D^2} \cdot e^{-\mu D} \, dD$$

which, on cancelling the common factors and integrating becomes:

$$N_{Max} = \underset{Lt\ D\to\infty}{N} = \underset{D\to\infty}{} \frac{\varepsilon\rho\pi a^2}{\mu} (1 - e^{-\mu D}) = \frac{\varepsilon\rho\pi a^2}{\mu} \qquad \text{(III,}$$

The two asymptotes bound the curves of the results for any energy and for any detector radius.

The graphs, one for each detector radius from 0.1, 1.0, 2.5, 5, 10 and 15 centimeter spheres, show the results for a set of 5 energies between energies 0.137 and 3.6 Mev. Useful interpolations can be made for other energies of interest.

The magnitudes of the responses of the detectors of increasing size may best be seen by noting the scales on the graphs for numbers of gamma crystal hits per unit volume activity and efficiency. These range from 1 for the maximum count rate with the 0.1 cm radius crystal to several times 10^4 with the 15 cm radius crystal. Making allowance for other than unit gamma emitter per unit volume and unit intrinsic efficiency will of course shift these magnitudes greatly. The scale for the abscissae extends to about 100 cm for all the energies considered.

The principal purpose of the work described has been to determine the increase in counting rate with increasing radii of crystal spherical detectors. The main effect of increasing the crystal size is of course to increase the solid angle of acceptance to gamma rays from isotopes in the neighborhood of the detector. Another effect occurs however, namely that of increasing the volume of water sampled with the detector; if mapping a region in seawater in searching for a source of activation is important the largest possible volume sample is desired. Increasing the counting rate by having the detector subtend a larger solid angle, is discussed first and a quantitative relation is given for the ratio of counts of a particular gamma ray with spheres of any two radii. Next the effect of increasing the volume sampled with larger detectors is shown to be relatively large but leads to a negligible increase in counting rate.

Before taking up the applications of the set of detectors the usefulness of equation (III) is examined in estimating the maximum counting rate for any size detector, or effective size detector, which is sampling a large body of seawater for the presence of a particular isotope. Evidently in formula (III) when ε, ρ and μ are constant the maximum counting rate depends only on the square of the detector radius. That is, for two detectors of radii in the ratio of a_1/a_2 the maximum counting rates are in the ratio of $(a_1/a_2)^2$. Relation (III) holds for any constant set of quantities ε, ρ and μ, which are respectively the intrinsic crystal efficiency, the specific concentration of the isotope and the attenuation coefficient in seawater for the gamma ray from the isotope. The relation is not restricted of course to the set of responses plotted for the several discrete energies in the graphs but applies to the gamma ray from any isotope for which the data required are known. This is the important effect of increasing the solid angle of acceptance of the detector by increasing the size.

An example will make clear the effect of increasing the volume sampled by going to larger crystals. Suppose a 1 cm crystal to be replaced by a crystal of 15 cm radius. Because the crystal is sensitive to radiation from several mean free paths distant, this

change increases the volume of seawater being sampled, at various efficiencies with distance, by a considerable amount. For a 1 Mev gamma ray in water, considering attenuation complete at 75 cms, increasing the crystal radius to 15 cms effectively adds 15 cms to the distance before complete attenuation is achieved, and so increases the volume being sampled by about 70 percent. The counting rate is thus increased, but only by about 1 percent, since the rays from the outer 70 percent increased volume are still being attenuated through the free paths closer in.

This example may be generalized to show, approximately, the part of the total counting rate from a small detector sphere which is due to the contributions from each shell of seawater one mean free path thick. The procedure is to use the reciprocal of μ as incremental radii of the shells, and read from the graphs the increase in counting rate due to the successive shells, for the detector sizes given. The added contributions from these shells 1, 2, 3, . . mean free paths from the detector center raise the counting rate approximately to 0.63, 0.86, 0.95, 0.98, 0.99, . . of the counting rate with an infinitely thick shell, making a very slow rise with volume. Computations of the same kind can be made from equations (III) using the exponential factor with μD.

Making use of (III) allows the computation of the volume being sampled with any detector sphere; that is the maximum volume which can have any appreciable effect on the detector immersed in the seawater. From the exponential factor in the next to last statement leading to the limit which is the maximum counting rate, the exponent contains the product μD. When within 1 percent of the limit, μD must have the value 5. Hence D_{max} for the sampled volume is $5/\mu$, depending on the attenuation coefficient μ for the particular energy of gamma ray being used in the analysis. Then the volume of the sphere of seawater being sampled is $(4/3)\pi D^3 = (4/3)(5/\mu)^3$. This relation confirms the earlier statement that at higher energy (lower attenuation, lower μ) more seawater is sampled, but as shown just above, as the distance increases, at progressively lower efficiency. The conclusion is that increasing the volume sampled by increasing the detector size is very inefficient and that the increase in volume sampled with a detector must be of volume close-in to the detector.

Finally, although for some purposes it may be useful to use a single large sphere at the center of a sampled volume, for other requirements it may be desirable to use a number of smaller spheres set at several mean free paths for attenuation from each other to sample a larger volume. The ratio of counting rate with a single

sphere to the volume of the single crystal sphere, namely

$$\frac{\varepsilon \rho \pi a^2}{\mu} \Big/ \frac{4}{3} \pi a^3 = \frac{3}{4} \frac{\varepsilon \rho}{\mu a}$$

shows that the counting rate per unit mass or volume of crystal decreases with the detector radius. That is, the larger spheres beyond a thickness about two mean free paths for interaction with the incoming gamma ray do not increase the counting rate, at least for low and medium energies, the reason being that beyond two mean free paths in the crystal the residual intensity has fallen greatly, leaving an ineffective crystal volume inside. Some work here and study of the literature has shown that the counting rates for crystal detectors vary with the surface area but become nonlinear with volume. (Private communication E. J. Wesley, NRDL.)

APPLICATION

A general example of the usefulness of this study will be given in showing how the response versus crystal sphere detector size would be used in attempting to reach the highest possible statistical accuracy in determining the presence of an isotope in the shortest time. The problem of determining the time since activation of the elements in seawater can be solved with efficient counting of two selected isotopes chosen by the methods outlined below.

Besides the roles of the detector size, and the intrinsic crystal efficiency, discussed above, the other factor in formula (III), namely the volume source strength of a gamma emitter, ρ_i, is important. The particular isotope, or isotopes, to be counted must be selected by a procedure such as is outlined below, in order to establish in a short time that the seawater has been activated at an earlier time.

The source strengths due to total number of atoms of each type of gamma emitter is computed as follows: The known concentration, C_o, of the inert isotope, i, which may be activated by a source of slow neutrons is multiplied by the activation cross section, σ_a, and the integral or fractional number, K, of gamma rays accompanying decay to the ground state with a characteristic mean lifetime τ_i. The initial

concentration of activated atoms of isotope i is then:

$$(C_o\ \sigma_a)_i\ \ \phi_o\ \ (\phi_o, \text{ neutron fluence}).$$

When multiplied by K this concentration is proportional to the initial source strength.

Since this isotope decays with mean lifetime, τ_i, the source strength at any later time, t, after activation varies as

$$(C_o\ \sigma_a\ K)_i\ \phi_o\ e^{-t/\tau_i}$$

When multiplied by the activity constant, $1/\tau_i$, this quantity gives the isotope gamma emission rate at any time after activation:

$$\rho_{i_t} = (C_o\ \sigma_a\ K)_i\ \phi_o\ e^{-t/\tau_i}\ \frac{1}{\tau_i} \qquad \text{(IV)}$$

To select the most likely candidate for analysis from a continuous or quasi-continuous distribution of isotopes in seawater (such as fission products) the maximum source strengths from activation would be arranged in order of their mean lifetimes. For a graphic display, and to aid selection, the maximum source strengths should be plotted on logarithmic paper with a logarithmic time scale. Then the first candidate to be considered would be the highest peak. If the mean lifetime were to be long, however, the activity constant $1/\tau_i$, could be small and make necessary a test of the next lower maximum source strength, i.e., (the next lower peak) for its suitability. Continuing in this way a decision would be made taking into account the factors affecting source strength, as discussed below, and then those affecting intrinsic efficiency, and the gamma ray energy, would be considered.

For a discrete distribution of isotopes in seawater the same procedure would be followed: isotopes missing from the continuous distribution discussed above would have no bearing on the selection, since only the highest peaks were considered, then the next highest, etc.

Having selected an isotope on the basis of source strength, it is evidently necessary to take into account the time of counting after activation as well as the mean lifetime of the candidate isotope.

The time since activation, t, will not usually be known, and may be the important datum being searched for if recency of activation is the information wanted. From the differential counting rates in the spectra for two different gamma peaks, separated by an interval of time however, the time since activation can usually be inferred.

To understand the effect of time of counting in attempting to reach the highest statistical accuracy consider the product in (IV) of the two factors $\frac{1}{\tau_i} e^{-t/\tau_i}$ on the selection of the particular isotope to count. This activity-constant-times-decay product evidently changes with time, but also with the selection of a particular isotope through the mean lifetime τ_i. It will always have its greatest value for a given isotope at the time of formation of the source, but if the mean lifetime of the isotope selected is too large the isotope will probably not be useful as was pointed out above.

Considered as forming a sequence of products in the order of increasing mean lifetimes (for a fixed real time after activation, t) the products for a <u>continuous</u> distribution of isotopes increase from zero at mean lifetime zero through a maximum at $\tau_i = t$, and back to zero at large τ_i. (The zero limit is shown at $\tau_i = 0$ by L'Hopital's Rule, and the occurrence of the maximum at $\tau_i = t$, by differentiation, as usual.)

For isotope i the maximum value of the source strength relative to all other isotopes formed at the same time in the series of products thus results for the values of (IV) when $t = \tau_i$.

$$\rho_i \tau_i = \frac{(C_o \sigma_a K)_i \phi_o}{e\tau_i}$$

This value of source strength is not the maximum the source has had but is in fact $1/e^{th}$ of this maximum. By waiting to count this source for a period equal to its mean life one sacrifices about two-thirds of the maximum counting rate but removes the activities having shorter mean lifetimes. For according to (IV) these activities will have been reduced to a few percent of their initial source strengths in two or three mean lifetimes. If the isotope considered has a sufficiently high initial activity according to the factor in parentheses (i.e., a high peak on the graph of selection) in (IV) it can be used to establish that the seawater has been activated.

A further factor which will be present whenever a collection of isotopes makes up a source is that the energies of the gamma rays emitted may or may not overlap in the spectrum, at any time. In general, following the above procedure of selection, using the mean lifetime order of counting the isotopes will give the maxima relatively separated in intensity, but not necessarily in energy, from the other simultaneously emitting isotopes. It may happen, however, that an intense line from some isotope will be well separated from all others in energy, and so allow an immediate count at the maximum intensity at zero time.

With the isotope or isotopes selected, making use of the successive graphs for the crystal sizes in Figures 2 to 7 (or alternatively formula (III)), would then allow an estimate of the necessary crystal size to reach a required statistical accuracy by registering a suitable number of counts within a required time.

SUMMARY AND CONCLUSIONS

An analysis has been made of the usefulness of increasing the size of a crystal detector immersed in seawater for determining, by spectral analysis, the concentration of gamma ray emitting isotopes, with the highest possible statistical accuracy in a short time. The counting rate with a crystal of arbitrary size is calculated, taking into account intrinsic efficiency of detection of a primary gamma ray in the crystal, the concentration of an isotope being sought, attenuation of the ray from the point of origin to the detector, and the solid angle of acceptance of the crystal assuming the volume of seawater containing the concentration is sufficiently large. The formulas developed can be used to analyze samples containing any emitter concentration in seawater for emitters having any intrinsic efficiency, and not just the unit concentrations and efficiencies graphed.

The results are given in the form of graphs giving counting rate for detectors of increasing size from 0.1 cm radius spheres to 15 cm radius, over energies from 0.137 Mev to 3.6 Mev gamma rays. The counting rates range from that of the smallest sphere at 1 count per second per unit volume activity and unit intrinsic efficiency to about 20,000 counts per second under the same conditions.

The increase of solid angle of acceptance is shown to be the important factor in increasing counting rate, with larger spheres, being in the ratio of the square of the crystal radii. The increased volume sampled is shown to be large but to yield a negligible increase in counting rate. Only the volume within the first two mean free paths for absorption in seawater is important in increasing the counting rate.

A relation (III) is given for the asymptotic maximum counting rate for spheres with any radii under any energy, in addition to the values given in the graphs.

A general example is given to show how the isotope to be counted would be selected from a continuous distribution of activated nuclei in seawater, either for maximum counting rate in a spectrum of gamma rays, or for recency of activation of the seawater. A suggestion is made for dealing with a discrete distribution of nuclei. The procedure described has a number of important aspects taken account of in formulas (III) and (IV), which must be kept in mind in selecting an isotope for gamma spectral analysis.

Use of the computations of response of the crystal spherical detectors given in the graphs is suggested in estimating the required size of crystal to reach a required statistical accuracy in counting an isotope within a specified time. Or the crystal size may be computed for gamma rays of any energy by formula (III).

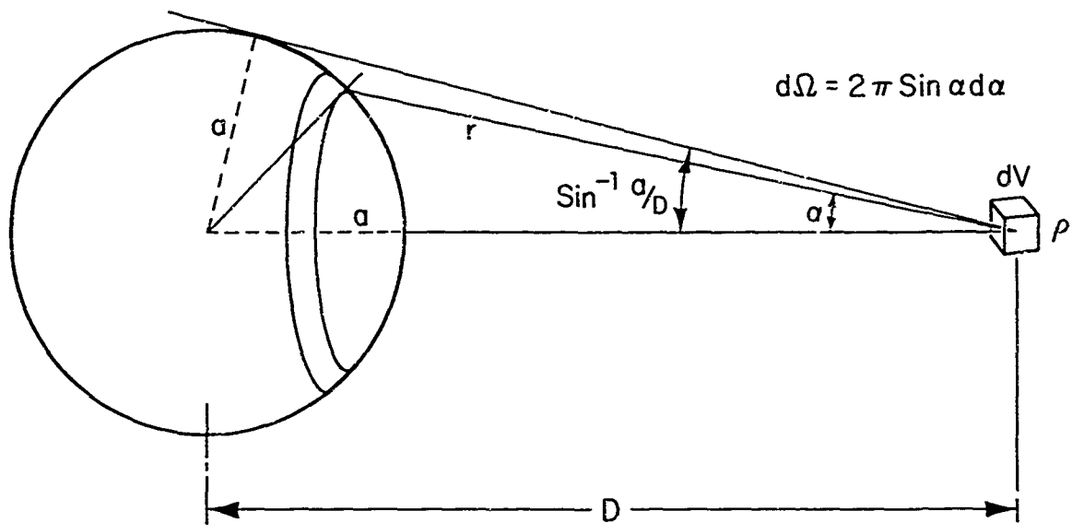

Fig. 1 Solid angle of acceptance on sphere.

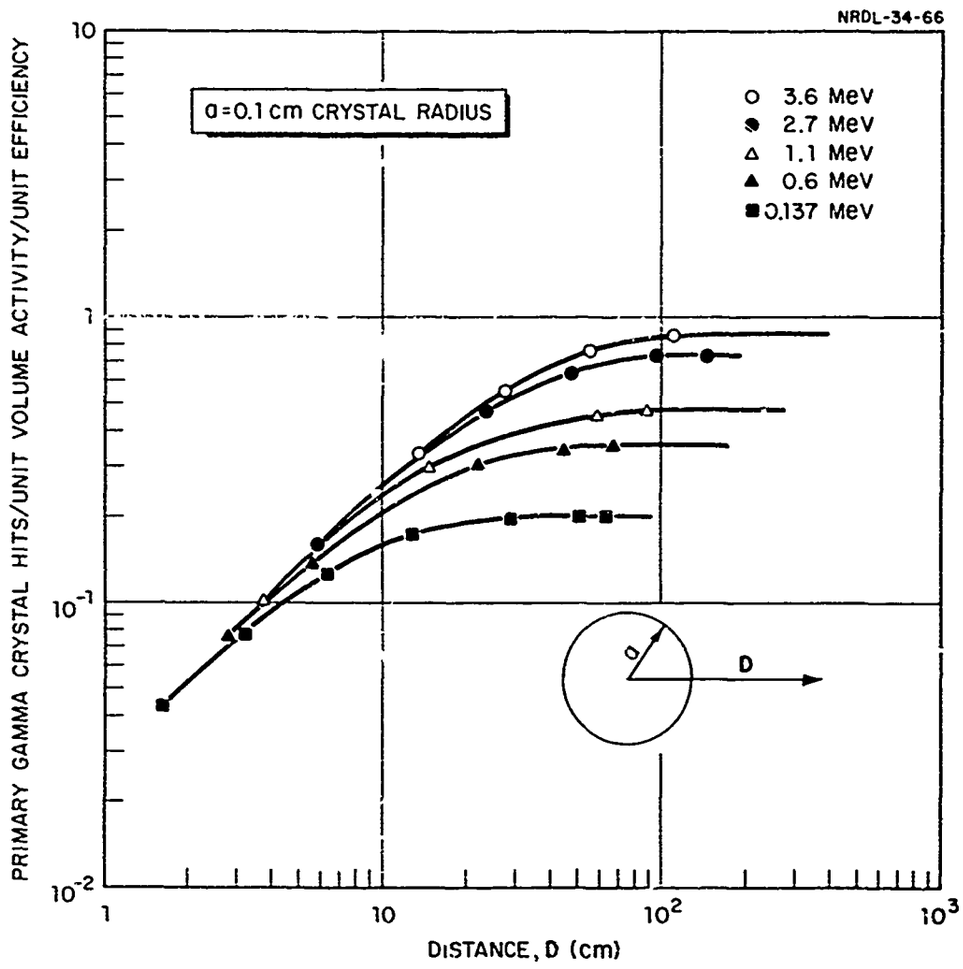

Fig. 2 Response of 0.1 cm radius crystal to gamma ray emitters at unit activity per unit volume, at unit efficiency.

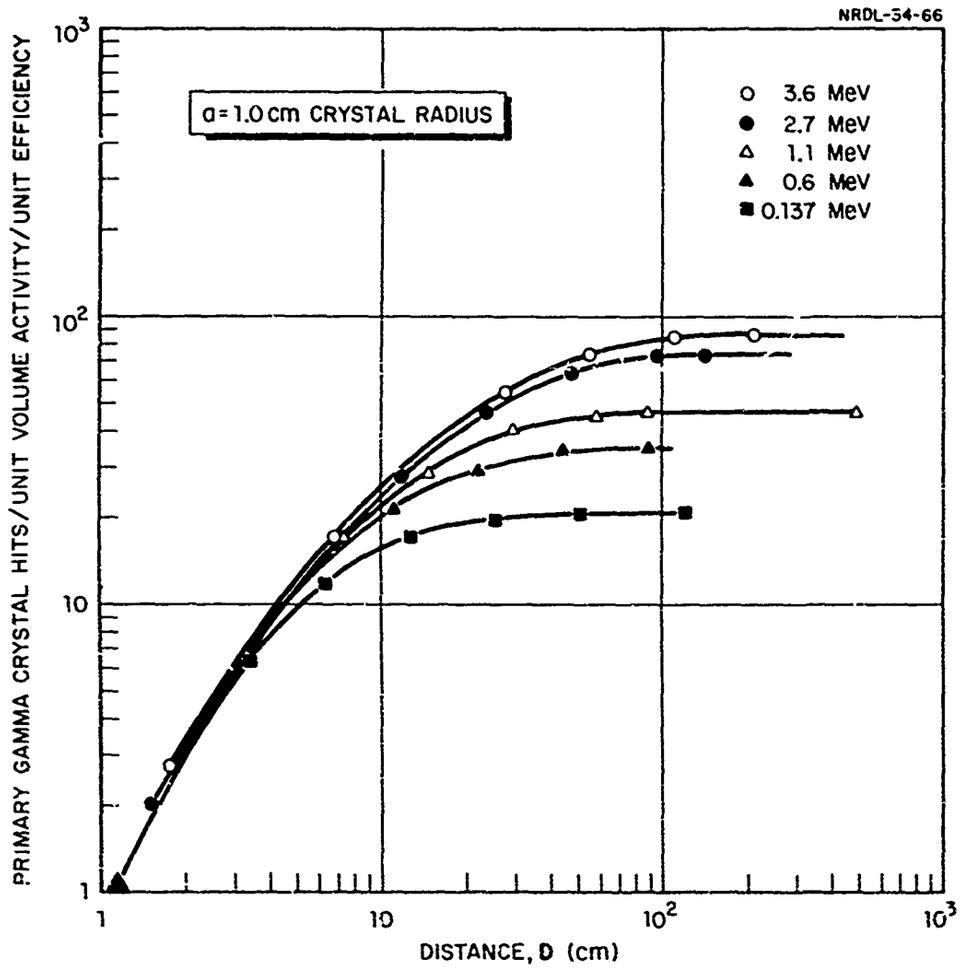

Fig. 3 Response of 1.0 cm radius crystal to gamma ray emitters at unit activity per unit volume, at unit efficiency.

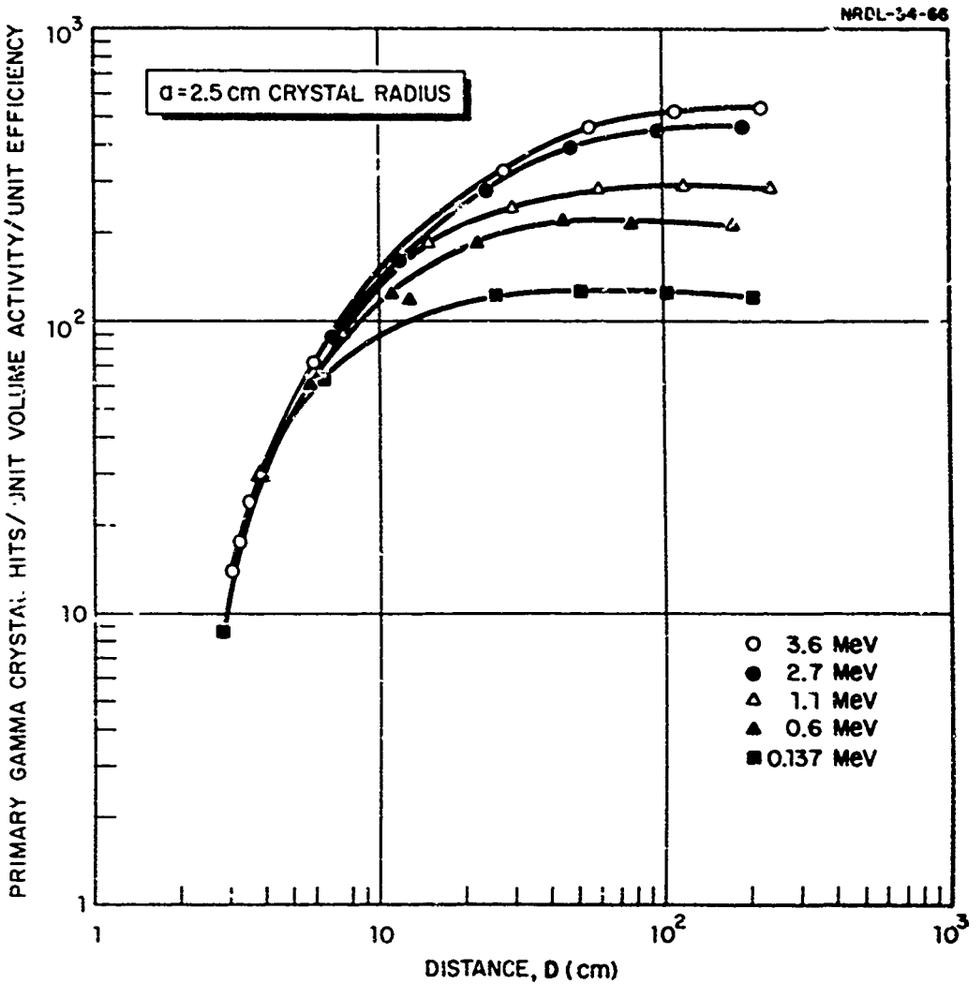

Fig. 4 Response of 2.5 cm radius crystal to gamma ray emitters at unit activity per unit volume, at unit efficiency.

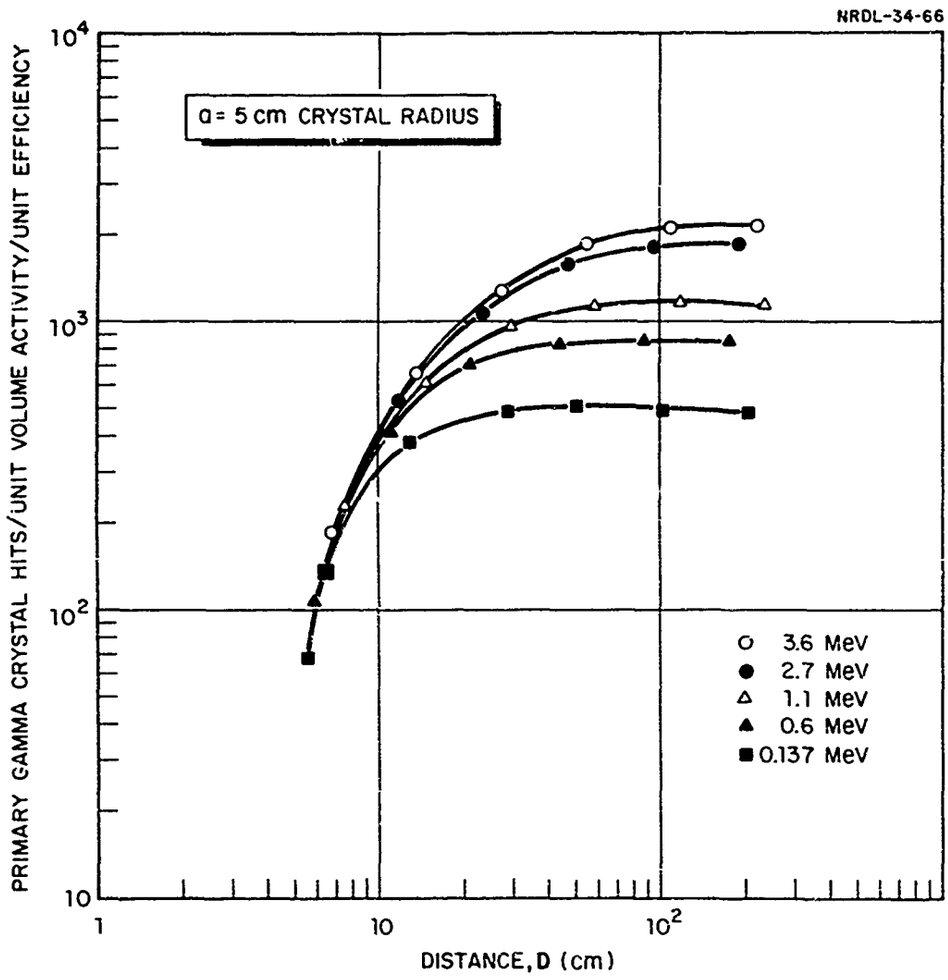

Fig. 5 Response of 5.0 cm radius crystal to gamma ray emitters at unit activity per unit volume, at unit efficiency.

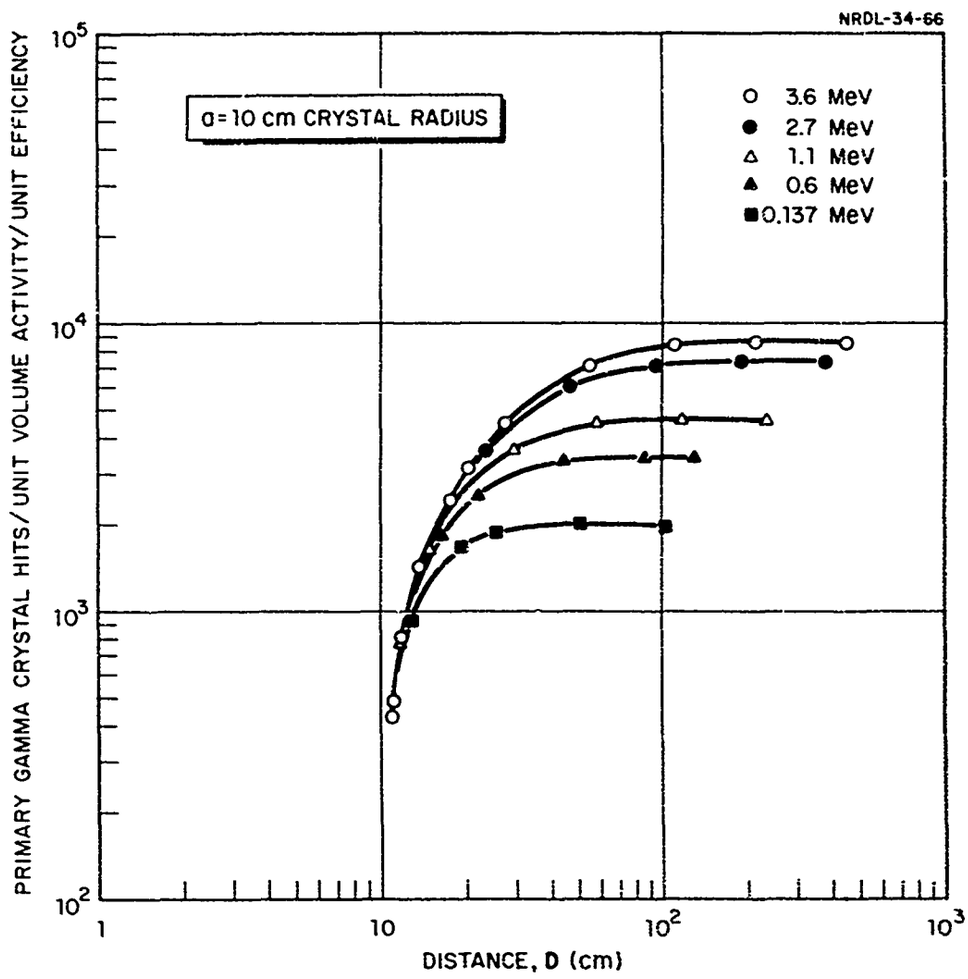

Fig. 6 Response of 10.0 cm radius crystal to gamma ray emitters at unit activity per unit volume, at unit efficiency.

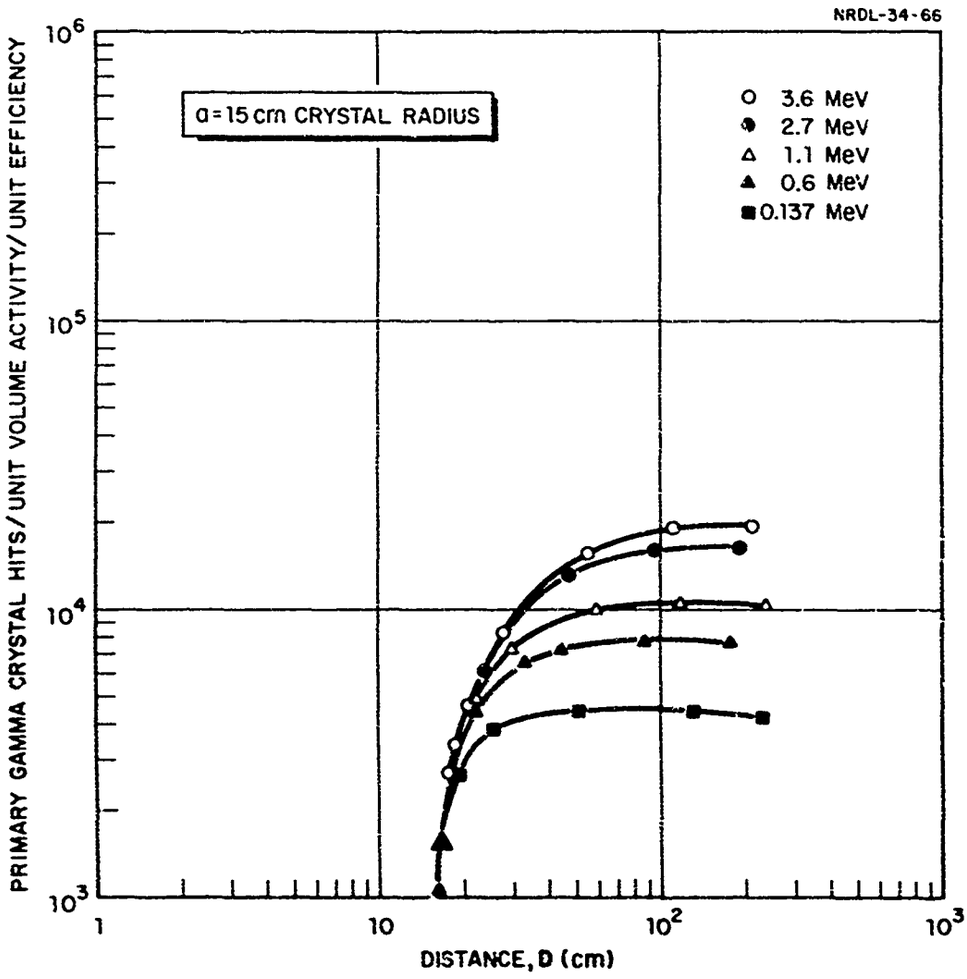

Fig. 7 Response of 15.0 cm radius crystal to gamma ray emitters at unit activity per unit volume, at unit efficiency.

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13. ABSTRACT The problem of assaying radioactivity in the sea has been under study with a view to designing an optimum detector for gamma radiation spectra, with suitable statistical accuracy.

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